07/714441

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION TO REISSUE :
U.S. PAT. NO. 4,912,155, :
ISSUED MARCH 27, 1990 :
:
SERIAL NO. :
:
FILED JUNE _____, 1991 :
:
FOR ANTIOXIDANT AROMATIC FLUORO- :
PHOSPHITES :

"Express Mail" mailing label number B89910190
Date of Deposit JUNE 13, 1991
I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, D.C. 20231
DEBORAH REAGAN
(Typed or printed name of person mailing paper or fee)
Deborah Reagan
(Signature of person mailing paper or fee)

INFORMATION DISCLOSURE STATEMENT UNDER 37 C.F.R. §1.97

Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

Pursuant to 37 C.F.R. §1.97, references which may be relevant to the examination of this reissue application are cited on the attached Form PTO-1449, and copies of the cited references are enclosed herewith. No representation is made that a search of prior art has been carried out or that no more pertinent prior art exists. However, the enclosed information represents the most relevant presently known information to applicant, his assignee, and undersigned counsel.

A brief description of the cited documents is set forth below. The descriptions are believed to be true and complete, but no representation is made, and applicant encourages the Examiner to review the cited art.

U.S.S.R. Authorship Certificate 787,412, published January 4, 1979, and Certificate 859,369, published January 15, 1980 (Vershinin):

These patents disclose chloro-analogs of the antioxidants of claim 1, formula II and chloro-analog of the compound of claim 5 of the invention in particular. None of the chloro-analogs of these references, however, are cited as antioxidants, but all are cited as intermediates in the production of other phosphite antioxidants (none of which are fluorophosphites). Similar references include the following, and show chloro-analog intermediates for other antioxidants:

U.S. Pat. No. 4,136,041, issued January 23, 1979 (Lenack) (See Example 1);

U.S. Pat. No. 4,259,492, issued March 31, 1981 (Rasberger)(see Cols. 7-8);

U.S. Pat. No. 4,374,219, issued February 15, 1983 (Spivack)(see Cols. 7-8 and Example A);

Canadian Pat. No. 1,159,078, issued December 20, 1983 (Spivack)(see pages 8-9);

Odorisio, Pastor and Spivack, "Reaction of Seven-and Eight-Membered Cyclic Phosphorochloridites With Alkanalomines", Phosphorus and Sulfur, 1984, Vol. 19 pp. 1-10; and

Odorisio, Pastor, Spivack and Steinhuebel, "12H-DIBENZO[d,g][1,3,2]DIOXAPHOSPHOCINS: Synthesis and Evidence for Long-Range Coupling to Phosphorus", Phosphorus and Sulfur, 1983, Vol. 15, pp. 9-13;

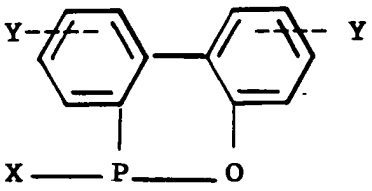
or other products:

U.S. Pat. No. 4,032,602, issued June 28, 1977 (Mazour) (intermediate for pest-control agent); and

U.S. Pat. No. 3,014,950, issued December 26, 1961 (Birum).

U.S. Pat. No. 3,702,878, issued November 14, 1972 (Saito):

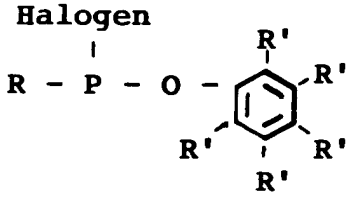
This reference discloses various tri- and penta-valent organophosphorus compounds, among which are included:



where X may be, among many other substituents, a halogen. However, the reference specifically discloses only chlorine and bromine. Flourine is never mentioned. There is no utility disclosed for these halogen (Cl and Br) or for any other compositions, although the reference generally states that "an object of the present invention [is] to provide new organophosphorus compounds useful as insecticides, fungicides, flame-retardants, antioxidants for organic compounds and so forth" (Col. 2, lines 15-18).

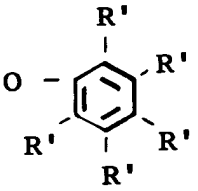
U.S.S.R. Authorship Certificate 398,574, published September 27, 1973 (Kim):

This reference discloses compounds of the formula:



where Halogen is F, Cl, Br or I; R' are C_1 to C_{10} alkyls; and

R is Halogen or - O -



These compounds are then reacted with polyphenylene ethers in the presence of metal alcoholate salts or amines to stabilize the polymers against high temperature. Although the general formula mentions that Halogen includes F, Cl, Br or I, only a chlorine analog is exemplified. With respect to the reference:

a) There is no enabling disclosure on how to make fluorophosphite analogs;

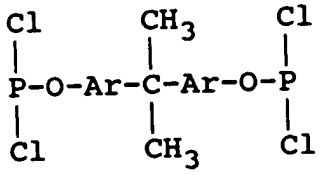
b) Fluorophosphites will not work as provided in the reference because the P-F bond will not be readily displaced by the polyphenylene ether;

c) Steric hinderance will preclude large or branched alkyl groups on the phenyl ring and there is no enabling disclosure for R' to be sec- or tert-alkyl; and

d) The reference teaches a reactive compound, not an additive to be incorporated into the organic material.

U.S. Pat. No. 3,254,050, issued May 31, 1966 (Baranauckas):

This reference discloses compounds of the generic formula:

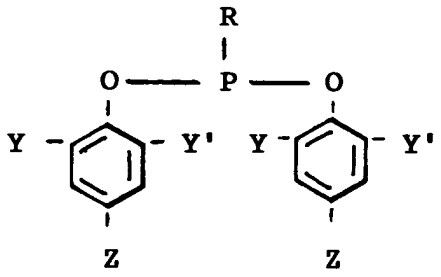


Ar is defined as an aromatic nucleus which can carry certain substituents (Col. 1, line 72 - Col. 2, line 3), but only phenylene

is exemplified. The disclosed compounds are said to be useful as heat stabilizers for vinyl and other resins and flame retardants for other polymer compositions (col. 1, lines 23-26). The patent states that "the chlorine may be substituted by another halogen" (Col. 1, line 38), but does not exemplify any other halogen, and does not suggest any utility for such modified product. Further, the reference does not enable the making of any fluorophosphite. The reference discloses that the compounds are made by reaction of bis-phenol with PCl_3. This method will not work with PF_3 because PF_3 is extremely toxic, has a boiling point of -101°C, and is not very reactive. There is no real suggestion and no incentive disclosed in the reference to modify the disclosure to make a fluorophosphite product, and no suggestion that such fluorophosphite would have any utility.

U.S. Pat. No. 3,281,506, issued October 25, 1966 (Shepard):

This patent discloses compounds of the formula:

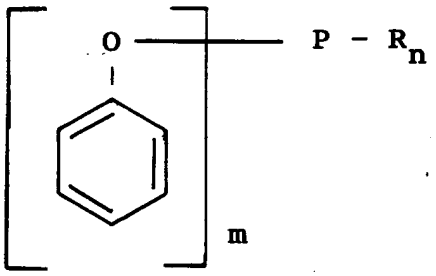


where R is hydroxyl or halogen, Y and Y' contain at least 4 carbon atoms, and Z can be hydrogen, halogen, or an organic radical. It is stated that the "secondary aryl phosphites" - - i.e., where R = OH, may be used as antioxidants, stabilizers, plasticizers, fuel

and oil additives, and flame retardants. There is no disclosure of any utility for the secondary aryl phosphoro-halidites, except as intermediates for the making of the "stable secondary aryl phosphite" (Col. 1, lines 56-62). The reference teaches the reaction of a phenol and PCl_3, and suggests that the PCl_3 may be substituted by PF_3 (Col. 4, line 21). No utility for such resulting product is disclosed, and there is no enablement for the use of the PF_3 mentioned in the reference. Unlike PCl_3, PF_3 is extremely toxic, has a boiling point of -101°C, and is relatively unreactive. There is no real suggestion and no incentive disclosed in the reference to make a fluoro-analog of the disclosed phosphite, and no utility is suggested for such product.

U.S. Pat. No. 4,094,855, issued June 13, 1978 (Spivack):

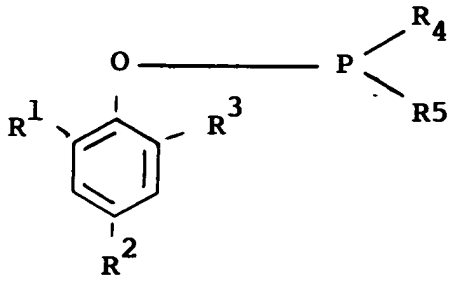
This patent was cited during the original prosecution. It discloses a phosphite of the formula:



where R can be, among many other substituents, a halogen, and m and n are 1 or 2 and m + n = 3. Flourine is never mentioned in this reference, and the only halogen exemplified is chlorine. It is stated that the compounds are light stabilizers, and "some of the phosphites of this invention act as antioxidants for polymers" (Col. 1, lines 42-44).

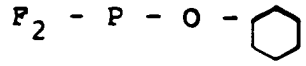
U.S. Pat. No. 4,233,207, issued November 11, 1980 (Spivack):

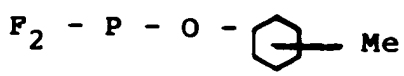
This patent was also cited during the original prosecution. It discloses compounds of the formula:

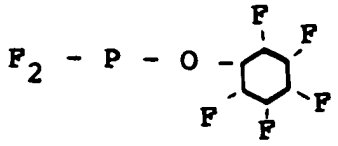


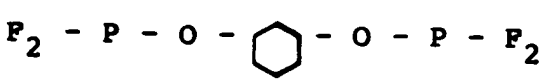
wherein R_1 and R_2 are alkyl, R_3 is hydrogen or alkyl, R_4 is alkyl, phenyl, or phenylalkyl, and R_5 can be, among many other substituents, a halogen. Halogen is chlorine or bromine (col. 3, line 65); fluorine is never mentioned. The components are generally reported to be useful as stabilizers, but none of the patent's utility examples shows any halogen-containing material. Thus, there is no specific utility cited for the disclosed materials where R_5 is a halogen.

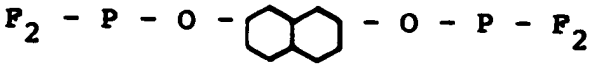
Below are set forth aromatic fluorophosphite structures which are disclosed in one or more of the references which are identified below. All such disclosures are to the bare compound, with no utility provided for any of the compounds. The compounds are:

1. 

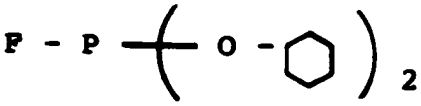
2. 

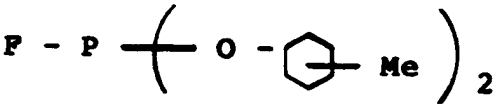
3. 

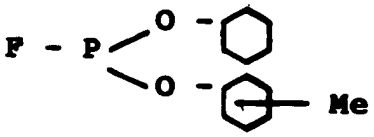
4. 

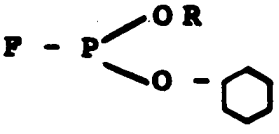
5. 

6. 

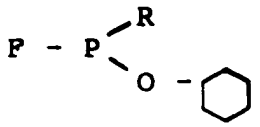
7. 

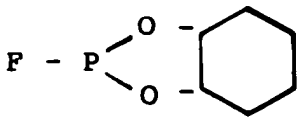
8. 

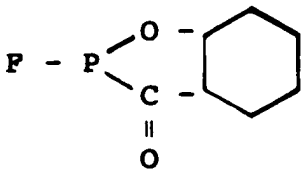
9. 

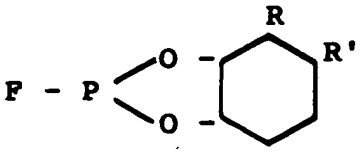
10. 

R = Bu, Me, i-Pr

11.  R = Me, Et

12. 

13. 

14. 

The references disclosing the above chemical structures are:

Kulakova, et. al., "Esters of Alkylfluorophosphonous Acids", J. of General Chemistry of the U.S.S.R., V. 39, No. 3, March 1969, pp. 547-49.

Razumova, et. al., "Condensation of Pyrocatechylphosphorous Acid Fluorides and Bromides with Diene Hydrocarbons", J. of General Chemistry of the U.S.S.R., V. 39, No. 1, January 1969, pp.162-66.

Razumova, et. al., "Glycolphosphorous Acid Fluorides", J. of General Chemistry of the U.S.S.R., V. 38, No. 5, May 1968, pp. 1072-76.

Ivanova, "Mono- and Difluorophosphites", J. of General Chemistry of the U.S.S.R., Vol. 34, No. 3, March 1964, pp. 852-55.

Schmutzler, "Synthese und Koordinationschemie der Fluorophosphite", Chemische Berichte, Germany, 1963, pp. 2435-50.

Ruppert, "Organyloxyfluorphosphorane: Direktsynthese durch F_2-Addition an Phosphinic-, Phosphonic- sowie Phosphoricsäure-ester(fluoride) und Thermolyseverhalten), Zeitschrift Für Anorganische Und Allgemeine Chemie, Germany, 1981, pp. 59-70

Micoud, et. al., "Étude magnétique (effet Faraday, aimantation, RMN) des complexes des types $Ni(P(OPh)_{3-x}F_x)_4$ et $Ni(PPh_{3-x}F_x)_4$,", Société Chimique de France, 1972, pp. 3774-82

Binder and Fischer, "Darstellung von Mono- und Difluorphosphiten", Zeitschrift Für Naturforschung, Germany, 1972, pp. 753-59.

Reddy and Schmutzler, "Nuclear Magnetic Resonance Studies on Fluorides of Trivalent Phosphorus" Zeitschrift Für Naturforschung, 206, 1965, pp. 104-09.

Schmutzler, "Phosphorus-Fluorine Chemistry. Part VIII. Some Comments on the 'Group Shift Theory', as Applied to the ^{31}P Nuclear Magnetic Resonance Spectra of Certain Phosphorus Fluorides." 1964, pp. 4551-57.

Kosolopoff and Maier, "Organic Phosphorous Compounds" Wiley-Interscience, New York, Vol. 5, pp. 148-49 and 292-95 (1973).

Respectfully submitted,

for [signature] Reg. No. 31,720
Rudolf E. Hutz
Registration No. 22,397
Telephone No. (302) 658-9141